THE

SCIENTIFIC PROCEEDINGS

OF THE

ROYAL DUBLIN SOCIETY

VOL. 25 (N.S.)

No. 23. DECEMBER 19, 1951.

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10 JAN 1952

DUBLIN :
PUBLISHED BY THE ROYAL DUBLIN SOCIETY,
BALL'S BRIDGE, DUBLIN

1951

Price Two Shillings

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THE THEORY OF THE USE OF ALPHA RAY RANGES IN NUCLEAR EMULSIONS FOR THE DETERMINATION OF THE RADIOACTIVE CONTENTS OF MATERIALS.

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[Read November 27, 1951. Published December 19, 1951].

THE estimation of the gross radioactivity of natural materials by the measurement of the rate of alpha ray emission from the surface of the material, is now a well known method. The original experiments on these lines were carried out by Evans and Finney (1) in 1935. They used an ionisation chamber, connected to a high gain amplifier and a photographically recording oscillograph, to count the ionisation pulses due to the individual alpha rays. Evans and Goodman (2), Nogami and Hurley (3), and Hurley (4), have employed similar methods since that date.

The development of nuclear emulsions in recent years has, however, rendered the actual technique of counting the alpha rays emitted from a surface far simpler. Since exposures up to about two months can be given, a much smaller rate of emission can be dealt with, and hence a much smaller area of specimen is required than in the ionisation chamber method. The nuclear emulsion method was originally proposed by Madame I. Joliot-Curie (5) in 1946, and has since been used by E. E. Picciotto (6), M. R. Coppens (7), and others.

Both methods, however, only measure the total alpha ray emission from the surface of the specimen, and a further relation is required to determine the absolute uranium and thorium contents. Madame Joliot-Curie originally proposed to obtain this extra condition by determining the ratio of the number of tracks in the emulsion, whose range exceeded that of Ra.C′. to the number of tracks, whose range lay between that of Ra.C′. and Th.A. E. E. Picciotto and M. R. Coppens have used this method. It is a very laborious one, since in determining the actual length of a track both its horizontal projection has to be measured with a micrometer eyepiece, and its vertical dip in the emulsion with the vertical slow motion of the microscope. This process involves four separate readings. Furthermore the measured dip has to be corrected for the shrinkage of the emulsion during processing, and also for the difference in refractive index between the medium in contact with the objective and emulsion, and the refractive index of the latter. The correction for the shrinkage of the emulsion is troublesome. Thus J. M. McAlister and D. W. Keam (8) find that not only does the shrinkage factor for a single plate vary appreciably with its hygrometric state, but also that this factor may vary much more between plates from different batches. It seems, therefore, worth considering, whether it would be possible to use the horizontal ranges only for the determination of the thorium to uranium ratio, since these ranges will not be affected by the shrinkage of the emulsion.

Two simple cases can be treated mathematically:

(a) A thin source, i.e. a plane surface distribution of uniformly radioactive material of neglible thickness compared to the range of the shortest alpha ray concerned;

(b) A thick source, i.e. a plane distribution of uniformly radioactive material of thickness greater than the longest alpha ray concerned.

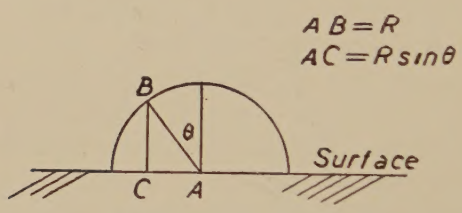
We will consider the problem of a single alpha ray emitter, say polonium, first.

Distribution of Horizontal Ranges from a Thin Source of a Single Alpha Ray Emitter.

Let N be the total number of alpha particles emitted per cm^2. of surface during the exposure time of the plate. Then the total number of tracks per cm^2. in the emulsion will be $\frac{1}{2}$ N.

Let P be the percentage of these tracks, whose horizontal range exceeds aR, where R is the maximum range of the alpha ray in the emulsion, and a is a proper

FIG. 1.



fraction. The chance that an alpha ray emitted at A (Fig. 1) makes an angle, with the normal to the surface, between θ and $\theta+d\theta$ is $\frac{1}{2}\sin\theta\, d\theta$. For alpha rays whose horizontal range is greater than aR, θ must lie between $\sin^{-1}a$ and $\frac{1}{2}\pi$. Hence the total number of such tracks is:

$$\tfrac{1}{2}\,N\int_{\sin^{-1}a.}^{\pi/2}\sin\theta.\,d\theta=\tfrac{1}{2}\,N\ \sqrt{1-a^2}.$$

Thus P is equal to $100\ \sqrt{1-a.^2}$

Table 1 shows the relative values of a and P obtained from this expression.

Table 1.

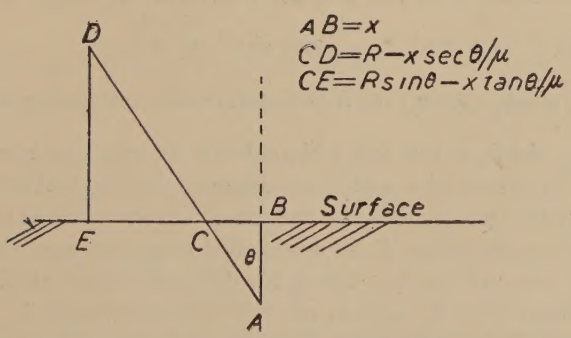
a	P. Thin Source	P. Thick Source
1.0	0	0
0.9	43.6	1.1
0.8	60.0	3.8
0.7	71.4	9.1
0.6	80.0	16.0
0.5	86.6	24.5
0.4	91.6	36.0
0.3	95.4	49.
0.2	98.0	62.
0.1	99.5	79.
0.0	100.	100.

Distribution of Horizontal Ranges from a Thick Source of a Single Alpha Ray Emitter.

Let N now be equal to the number of alpha particles emitted per c.c. of the source during the exposure time, R the maximum range in air, and μ the ratio of the range in the source to the range in air. We will consider the distribution of the horizontal ranges of the alpha particles in the air over the source, since the same percentage distribution will apply to the emulsion.

Consider an alpha particle emitted at a depth x below the surface of the source (Fig. 2). The chance that it is emitted at an angle between θ and $\theta+d\theta$ is, as before

FIG. 2



$\frac{1}{2} \sin\theta . d\theta$. Its residual range in air will be R—x sec θ/μ, and its horizontal range in air will be R sin θ—x tan θ/μ.

$$\text{Let } x = R\mu k$$
$$\text{so that } dx = R\mu dk$$

and let n be the number of tracks per cm². whose horizontal range exceeds aR. Now as θ decreases from $\pi/2$ to o, the horizontal range for a given value of x, will increase from zero to a maximum, and then decrease to zero again. It will exceed aR between values θ_1 and θ_2 where these are the real roots of the equation :

$$aR = R\sin\theta - x\tan\theta/\mu$$
$$\text{or } a = \sin\theta - k\tan\theta.$$

The chance that an alpha particle is emitted at an angle between θ_1 and θ_2 is $\frac{1}{2}\int_{\theta_1}^{\theta_2} \sin\theta d\theta$, or $\frac{1}{2}(\cos\theta_1 - \cos\theta_2)$.

$$\text{Hence } dn = \tfrac{1}{2}(\cos\theta_1 - \cos\theta_2)\, N\, dx$$
$$= \tfrac{1}{2} N R \mu (\cos\theta_1 - \cos\theta_2) dk$$
$$\text{Thus } n = \tfrac{1}{2} N R \mu \int_0^K (\cos\theta_1 - \cos\theta_2)\, dk$$

If we put a = o, we get the total number of tracks emitted per cm². of surface. The limits of the integral are then o and 1, and $\cos\theta_1 - \cos\theta_2 = 1 - k$. Hence the total number of tracks is N $R\mu/4$, a well known result. Thus finally P, the percentage of tracks whose horizontal range exceeds a R is $200 \int_0^K (\cos\theta_1 - \cos\theta_2)\, dk$.

The value of the upper limit K of this integral can be easily found. If k is kept constant, the maximum horizontal range is given by the condition:

$$\frac{d}{d\theta}(\sin\theta - k\tan\theta) = o, \text{ or } k = \cos^3\theta.$$

For this value of k :

$$a = \sin\theta - k\tan\theta = \sin\theta(1 - \cos^2\theta) = \sin^3\theta$$

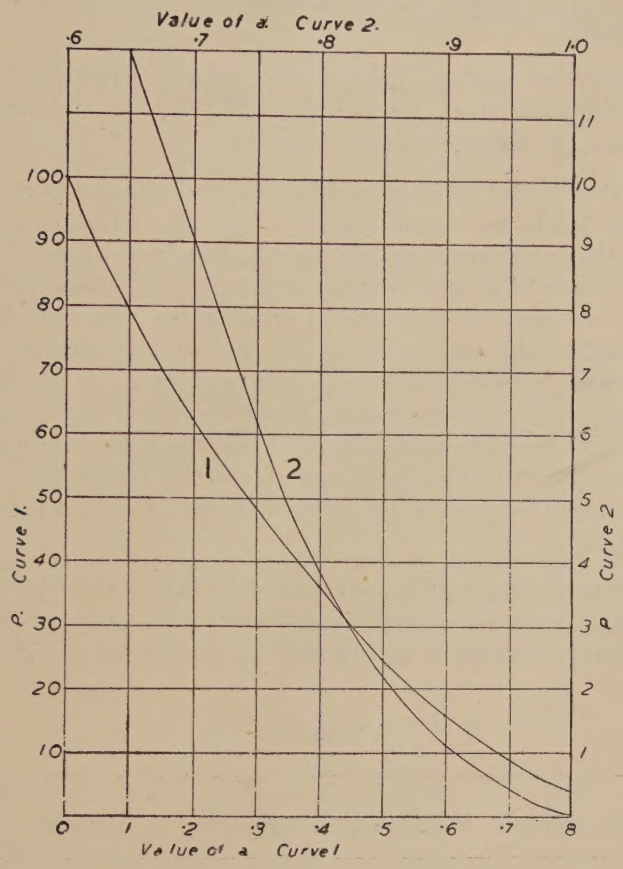
Thus $\theta = \sin^{-1}\sqrt[3]{a}$ and $K = \left(\cos\sin^{-1}\sqrt[3]{a}\right)^3$

To evaluate $\int_o^K (\cos\theta_1 - \cos\theta_2)\, dk$ it is necessary to use approximate methods.

The equation, a = sin θ—k tan θ is a biquadratic for sin θ, and certainly not easily soluble. If definite values of a and k are assumed its two real roots, however, can be obtained with sufficient accuracy by the use of trigonometrical tables. Supposing therefore we wish to determine P for horizontal ranges greater than, say, o. 7 R, we calculate the values of cos θ_1—cos θ_2 for different values of k, keeping a equal to o·7. The values of cos θ_1—cos θ_2 are next plotted against k, and the required integral found from the area included between this curve and the axis. The points at which this curve cuts the axis are known, since for k = o, cos θ_1—cos θ_2 = $\sqrt{1-a^2}$, and for cos θ_1—cos θ_2 = O, k = K = (cos $\sin^{-1}\sqrt[3]{a})^3$. This process can

now be repeated for various values of a. The results are given in Table 1 and they are also shown graphically in Fig. 3.

FIG. 3.



Determination of the Ratio of the Thorium to Uranium Contents from the Distribution of the Horizontal Ranges.

In the following discussion it will be assumed that the thorium and uranium families are in equilibrium, and the small effect due to the actinium or U.235. series will be neglected, since the accuracy is not great enough to justify its inclusion. The final formulae can be easily corrected for the effect of this series, if required. Two different methods may be used.

(1) The ratio of the number of tracks per cm^2., whose horizontal range exceeds the maximum range of Ra.C′ to the total number of tracks per cm^2. is determined and from this figure the thorium uranium ratio is found.

(2) The ratio of the number of tracks whose horizontal range exceeds the maximum range of Ra.C′ to the number of tracks, whose horizontal range lies between the maximum range of Ra.C′ and Th.A is determined. The thorium uranium ratio can then be found in a manner similar to Madame I. Joliot-Curie's original method.

Once the thorium uranium ratio has been found by either method, the absolute thorium and uranium contents can be calculated from the total alpha ray emission per cm^2. per sec. as determined from the plate.

Method (1) is bound to be unsatisfactory for a thick source, since for even a pure thorium family source only about 1 per cent of the tracks will exceed in horizontal length the maximum range of Ra.C′. Thus a very large number of tracks will have to be counted to give even very approximate results. For a thin source the position is not quite so bad, but, in practice, the total emission from a thin source of a weakly radioactive material may be too small, and the preparation of the source difficult. Method (2) is more hopeful, since it is only necessary to measure tracks in any one field of the microscope whose horizontal length exceeds a definite quantity, and, with practice, such tracks can be usually identified by mere inspection. It should be noted that this method gives the thorium radium ratio correctly, even if some of the upper members of the uranium series are not present in their equilibrium amounts.

The theory of the two methods, as applied to both thin and thick sources, follows. For ease of reference the various alpha ray ranges for the three naturally radioactive series, as given by Madame I. Joliot-Curie (5), are shown in Table 2.

TABLE 2.

Element	Range	Element	Range	Element	Range
U.238	2.65 cm.	U.235	2.82 cm.	Th.232	2.60 cm.
U.234	3.21 cm.	Pa.	3.51 cm.	Ra.Th	3.98 cm.
Ionium	3.11 cm.	R.Ac	4.72 cm.	Th.X	4.28 cm.
Radium	3.30 cm.	Ac.X	4.32 cm.	Thoron	5.00 cm.
Radon	4.05 cm.	An	5.69 cm.	Th.A	5.64 cm.
Ra.A	4.66 cm.	Ac.A	6.46 cm.	Th.C 35%	4.66 cm.
Ra.C′	6.91 cm.	Ac.C	5.43 cm.	Th.C′ 65%	8.57 cm.
Polonium	3.84 cm.				

The total emission from the complete uranium series under equilibrium conditions is eight times that of U.238, whereas for the Thorium series it is six times that of Th.232.

Method 1.

(*a*) *Thin Source.*

Let, N_u = the number of alpha particles emitted per sec. per g. of U.238 = 1.2×10^4

N_{th} = the same quantity for Th.232 = 0.4×10^4

W_u = surface density in g./cm². of U.238

W_{th} = ditto for Th.232.

Then the total emission per cm². of surface per sec. is equal to

$$\tfrac{1}{2}\ (8\ N_u\ W_u + 6\ N_{th} W_{th}).$$

The total number of alpha particles per sec., derived from the actual count on the plate will be slightly less than this figure, owing to the fact that tracks, whose horizontal range is less than a certain length, will not be counted. From actual measurements this seems to correspond to a range of 8 mm. in air. Thus the counted emission from any one member of the uranium series will be $\frac{1}{2}\sqrt{1-a^2}\ N_u\ W_u$, where a is the ratio of 8 mm. to the range in air of the alpha particle concerned. A similar correction can be applied to the thorium series, allowance being made for the branching of the series at Th.C. The branching of the uranium series at Ra.C. need not be considered, since Ra.C′. is produced in 99.96 per cent of its disintegrations. Thus if p_1 is the total alpha ray emission rate obtained from the plate count, we find that ;

$$p_1 = \tfrac{1}{2}\ (7.78\ N_u\ W_u + 5.88\ N_{th}\ W_{th}).$$

The total emission rate due to Th.C′. only is $\frac{1}{2}(0.65\ N_{th}\ W_{th})$. Let p_2 be the number of these alpha rays whose horizontal range exceeds the maximum range of Ra.C′.

$$\text{Then } p_2 = \tfrac{1}{2}\ (0.65\ N_{th}\ W_{th}\sqrt{1-a^2}).$$
$$\text{where } a = 6.91/8.57 = .806.$$

$$\text{This gives } p_2 = \tfrac{1}{2}\ (0.38\ N_{th}\ W_{th}).$$

$$\text{Thus } p_1/p_2 = 20.4\ N_u\ W_u/N_{th}\ W_{th} + 15.5$$

$$\text{or finally } W_{th}/W_u = 61/(p_1/p_2 - 15.5).$$

Hence if W_{th}/W_u is about 3, the value for an average acid igneous rock ; $p_1 = 36\ p_2$, so that only about 3 per cent of the total tracks counted would be worth measuring. It follows that the statistical error in p_2 will be unduly high, unless a very large number of tracks are counted.

(b) Thick Source.

Let C_u = uranium content of the source in g./g.
C_{th} = thorium content of the source in g./g.
d = density of the source in g./c.c.
μ = the ratio of the alpha ray range in the source to their range in air.

and p_1 and p_2 have the same significance as before.
The total emission per cm². per sec. of alpha rays from any one member of the uranium series is $\frac{1}{4}\mu N_u d C_u R$, where R is the range of the alpha particle concerned. Only a certain fraction of these alpha particles will have horizontal ranges exceeding 8 mm. The value of this fraction for any given member of the series can be determined from Fig. 3. When this is done for all the members of both series, and the necessary summation performed; we find that,

$$p_1 = \tfrac{1}{4}\mu\,(19{\cdot}9\ N_u\ C_u + 19{\cdot}5\ N_{th}\ C_{th})\ d.$$

The total emission per cm². per sec. from Th.C′ is equal to,

$$\tfrac{1}{4}\mu \times 8{\cdot}57 \times 0{\cdot}65\ N_{th}\ C_{th}\ d = \tfrac{1}{4}\mu \times 5{\cdot}56\ N_{th}\ C_{th}\ d.$$

The percentage of these alpha particles, whose horizontal range exceeds the maximum range of Ra.C′ is 3·6, as determined from Fig. 3.

Thus $p_2 = \frac{1}{4}\mu \times 0{\cdot}20\ N_{th}\ C_{th}\ d.$
and $p_1/p_2 = 5{\cdot}0\ (19{\cdot}9\ N_u\ C_u/N_{th}\ C_{th} + 19{\cdot}5).$
or finally, $C_{th}/C_u = 300/(p_1p_2 - 97).$

Thus for an average acid igneous rock, where C_{th}/C_u is about equal to 3, p_1/p_2 would be about 200, *i.e.* that only one track in two hundred would exceed in horizontal length the maximum range of Ra.C′. This method, accordingly, seems a very unsatisfactory one.

Method 2.

(a) Thin Source.

Let n_1 = the number of alpha particles per cm². per sec. whose horizontal range exceeds the maximum range of Ra.C′.
n_2 = the number of alpha particles per cm². per sec. whose horizontal range lies between the maximum ranges of Ra.C′ and Th.A.

Since we know the ratios of the maximum ranges of Th.C′, Ra.C′ and Th.A, we can calculate from the theory of the distribution of horizontal ranges from a thin source that,

$$n_1 = \tfrac{1}{2} \times 0{\cdot}38\ N_{th}\ W_{th}.$$
$$n_1 + n_2 = \tfrac{1}{2} \times (0{\cdot}49\ N_{th}\ W_{th} + 0{\cdot}57\ N_u\ W_u).$$

This gives $W_{th}/W_u = 4{\cdot}5/(n_2/n_1 - 0{\cdot}29).$
Thus for W_{th}/W_u 3, $= n_2 = 1{\cdot}79\ n_1.$

The method seems therefore quite feasible, provdied thin sources can be used, but this may be difficult to realise for weak sources.

(*b*) *Thick Source.*

The total number of alpha particles per cm². per sec. due to Ra.C′ is $\frac{1}{4}\mu \times 6.91\ N_u\ C_u\ d$, and the number from Th.C′ is $\frac{1}{4}\mu \times 5.56\ N_{th}\ C_{th}\ d$. From Fig. 3, we find that 3·4 per cent of the Ra.C′ alpha particles will exceed in horizontal range the maximum range of Th.A, while of the Th.C′ alpha particles 3·6 per cent will exceed the maximum range of Ra.C′, and 11·3 per cent that Th.A in horizontal length.

From this it follows that :

$$n_1 + n_2 = \tfrac{1}{4}\mu\ (0.63\ N_{th}\ C_{th} + 0.24\ N_u\ C_u)\ d$$

$$n_1 = \tfrac{1}{4}\mu \times 0.20\ N_{th}\ C_{th}\ d.$$

This gives :

$$C_{th}/C_u = 3.6/(n_2/n_1 - 2.15).$$

Hence for $C_{th}/C_u = 3$, $n_2 = 3.4\ n_1$.

It is of some interest to compare these expressions with those obtained from the original method, in which the total range of the alpha particle in the emulsion has to be measured. M.R. Coppens (7) using this method finds,

$$C_{th}/C_u = 3.3/(n_2/n_1 - 0.8).$$

where,

n_1 = the number of tracks of total length greater than 7·0 cm.

n_2 = the number of tracks of total length between 5·8 cm. and 7·0 cm.

and the small effect of the actinium series is allowed for. This gives $n_2 = 1.90\ n_1$, if $C_{th}/C_u = 3$. It will be seen that this method yields a more advantageous value for the ratio of n_2 to n_1, but this gain is probably outweighed by the difficulty of determining the total ranges accurately. Both methods can only give reliable results when the total number of tracks is sufficiently large, since only the longer tracks are used.

Experimental Results.

To test the validity of the various formulae obtained for the different cases considered, measurements on plates exposed to various types of source were performed.

Thin Source—Single Alpha Ray Emitter.

A thin source of polonium was prepared by auto-deposition on a nickel plate from a solution of RaD+E+F. We are indebted to C. F. G. Delaney for the preparation of this source, which was originally made in connection with the determination

of the beta disintegration constant of K.40. (9). Three plates were exposed to this source, the first immediately after the preparation of the source, the second 23 days later and the third 235 days later. The exposures were varied so that the number of tracks per field in the microscope were about the same for the three plates. The first two plates were exposed and processed by Delaney. The results of horizontal range measurements on the three plates are shown in Table 3. A Bausch and Lomb microscope and micrometer eyepiece were used, a 4mm. dry objective being employed. For the measurement of horizontal ranges only, a dry objective is convenient since its depth of focus is greater than that of an oil immersion objective, and any possible distortion of the emulsion due to the oil is avoided. The loss of magnification is not really serious.

TABLE 3.

a	P Theoretical	P Plate 1	P Plate 2	P Plate 3
0.9	43.6	41	28	22
0.8	60.0	58	56	44
0.7	71.4	68	66	56
0.6	80.0	79	73	63
0.5	86.6	85	81	69
0.4	91.6	91	86	76
0.3	95.4	94	91	89
0.2	98.0	97		
0.1	99.5	100		
0.0	100			

P = the percentage of tracks with horizontal range greater than a R, where R is the maximum horizontal of the polonium alpha ray in the emulsion. Number of tracks measured, Plate 1-300, Plate 2-600, Plate 3-200.

It will be seen that the agreement between the theoretical and experimental results is quite satisfactory for Plate 1, but that the two later plates show a progressive decrease in the number of longer tracks. This is probably due to the tarnishing of the source, so that it no longer behaves as a true surface deposit.

Thick Source.—Single Alpha Ray Emitter.

A thick source of polonium was prepared by the following method. A solution containing Ra.D+E+F was made by treating a few ground up old radon tubes with dilute HC1. The powdered glass was removed by filtration and a small quantity

of $PbNO_3$ added to the solution. Ammonium sulphate was added to the solution, and the lead precipitated as lead sulphate. Since Ra.D. is an isotope of lead it was precipitated in an uniform distribution throughout the inactive lead sulphate, and polonium (Ra.F.) will grow through its decay. It is likely that some Ra.E. and Ra.F were also precipitated, but it seems probable that they were uniformly distributed too. The lead sulphate was filtered out and dried, and a thick source made from the powder. A plate placed in contact with this source gave a horizontal range distribution as shown in Table 4.

Since the minimum visible horizontal range corresponds to about 8 mm. of air, or about 0·2R, where R is the range of a polonium alpha particle ; it was assumed that the theoretical and experimental results should coincide for a = 0·2. The other experimental results were corrected accordingly. It will be seen that some tracks of horizontal length between 0·2R and 0·1R were actually measured, but this is to be expected since the exact minimum length visible is uncertain, and will vary for different observers. It is not necessary to correct the results for the polonium thin source in this way, as the number of tracks of horizontal length less than 0·2R is very small.

It will be seen that the experimental results, when corrected agree as well as could be expected with the theoretical values. The statistical error in the percentages of the longer tracks is bound to be large since so few of these tracks occur.

Table 4.

a	P. Theory	P. Experimental	P. Corrected
0·9	1·1	2·0	1·4
0·8	3·8	7·5	5·2
0·7	9·1	14·	9·8
0·6	16·0	24·	16·9
0·5	24·5	36·	25·3
0·4	36·0	51·	35·8
0·3	49·0	70·	49·2
0·2	62·0	88·	62·
0·1	79·0	100	70·5
0	100		

Total number of tracks measured = 500.

Thick Source.—Family of Radioactive Elements.

Thorium oxide over thirty years old, so that all the members of the thorium family were present in practically equilibrium amounts, was used as a source. All tracks with horizontal ranges greater than the range of Th.A were measured, and the total number of tracks in each microscope field counted. Thus methods 1 and 2 could both be applied.

Method 1.

Total counted emission = 2029. Number of tracks with horizontal range greater than that of Th.A = 70.

Thus $p_2/p_1 = 70/2029 = 3{\cdot}45\%$.
Theoretical value $= 3{\cdot}2\%$.

Method 2.

Number of tracks with horizontal range greater than that of Ra.C′. = 23.

Number of tracks with horizontal ranges greater than the range of Th.A and less than that of Ra.C′. = 47.

Thus $n_2/n_1 = 47/23 = 2{\cdot}04\%$.
Theoretical value $= 2{\cdot}15\%$.

Hence the experimental results agree satisfactorily with the theoretical values for both methods.

Some experiments have also been made on natural materials, but the results obtained to date have been confusing. In most cases the number of long tracks has proved to be much too large. The thick source method has, so far, been employed, since the materials used were only feebly radioactive. The discrepancy must be due to the fact that the material does not act as a uniform thick source, even when finely powdered. This effect has already been observed by Hurley (4) and Poole, Delaney, and McCormick (10). In both these series of experiments the total ranges were measured. Further work on this problem is in hand.

Summary

The theory of the determination of the Thorium Content to Uranium Content Ratio for thin and thick radioactive sources by the measurement of the horizontal alpha ray ranges only in a nuclear emulsion, is given ; and some results, confirming the theoretical values, described. The difficulty of applying the thick source theory to actual natural materials is briefly discussed.

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